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MAGNETIZATION AND MAGNETIC ANISOTROPY OF TbFe2, DyFe2, Tb0.27 Dy0.73 Fe2 AND TmFe2

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Saturation magnetizations were measured on single crystals of TbFe<sub>2</sub>, DyFe<sub>2</sub>, Tb  $_{27}$ Dy<sub>0.73</sub>Fe<sub>2</sub> and TmFe<sub>2</sub>. Over most of the temperature range from 4 K to 300K, these values are substantially larger than those measured earlier on polycrystals. The intrinsic magnetic anisotropies, K<sub>1</sub> (0)'s, as determined from magnetic fields required for saturation, are huge ( $\sqrt{5} \times 10^{\circ}$  erg/cm). For these highly magnetostrictive compounds, the magnetoelastic contribution to the anisotropy,  $\sqrt{2.44 \times 111}$  is a significant fraction of the total anisotropy.

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## SUMMARY

In this report are presented saturation magnetization measurements on single crystal TbFe<sub>2</sub>, DyFe<sub>2</sub>, Tb<sub>0.27</sub>Dy<sub>0.73</sub>Fe<sub>2</sub> and TmFe<sub>2</sub>. These compounds are strongly magnetic, highly magnetostrictive, and possess a wide range of cubic magnetocrystalline anisotropy,  $K_1$  - ranging from the largest known (5 x 10<sup>8</sup> erg/cm<sup>3</sup>) for TbFe<sub>2</sub> to a small value (<10<sup>6</sup> erg/cm<sup>3</sup>) for the compensated Tb<sub>0.27</sub>Dy<sub>0.73</sub>Fe<sub>2</sub> compound. From the magnetization vs. field measurements along the principal axes, the magnetic anisotropy was calculated as a function of temperature for DyFe<sub>2</sub> and TmFe<sub>2</sub>. It was discovered that, even in the noncompensating compounds, the magnetoelastic contribution to the magnetic anisotropy ( $\Delta K_1 = -\frac{1}{2}C_{44} \lambda_{111}^2$ ) is a significant portion (~ 20%) of the intrinsic anisotropy constant,  $K_1$ . For the case of the compensated Tb<sub>0.27</sub>Dy<sub>0.73</sub>Fe<sub>2</sub> compound, the temperature dependence of the magnetomechanical coupling factor was correlated to the temperature dependence of the anisotropy field.

This study was carried out in the Solid State Branch of the Radiation Division as part of the research program on magnetostrictive materials. The research was sponsored by the Office of Naval Research (PO 4-0081, NR 039-110) and the NSWC Independent Research Funds.

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#### **ABSTRACT**

Saturation magnetizations were measured on single crystals of TbFe, DyFe, Tb, 70, 73Fe, and TmFe. Over most of the temperature range from 4 K to 300 K, these values are substantially larger than those measured earlier on polycrystals. The intrinsic magnetic anisotropies,  $K_1$  (0)'s, as determined from magnetic fields required for saturation, are huge ( $\sim$ 5 x 10°) cm3). For these highly magnetostrictive compounds, the magnetoelastic contribution to the anisotropy  $-\frac{1}{2}c_{44}\lambda_{111}^2$  is a significant fraction of the total anisotropy.

## INTRODUCTION

The rare earth-Fe<sub>2</sub> Laves phase compounds possess the largest known cubic anisotropies. At low temperatures, anisotropies of DyFe<sub>2</sub> and ErFe<sub>2</sub> exceed 5 x 10 erg/cm<sup>2</sup>; at room temperatures, anisotropies of TbFe<sub>2</sub> and DyFe<sub>2</sub> exceed 2 x 10 erg/cm<sup>2</sup>. The RFe<sub>2</sub> compounds also possess huge magnetostrictions, which also extend to room temperature for TbFe<sub>2</sub> and SmFe<sub>2</sub>. In this paper we report magnetization measurements on single crystals of TbFe<sub>3</sub>. DyFe<sub>4</sub> The DyPa = Fe<sub>4</sub> and TmFe<sub>4</sub>. From measurements of TbFe, DyFe, Tb<sub>0,27</sub>Dy<sub>0,73</sub>Fe, and TmFe, From measurements along the hard magnetic directions, magnetic anisotropy energies are calculated. In magnetostrictive materials, the magnetic anisotropy depends upon the state of strain-a clamped sample possessing the conventional intrinsic anisotropy, described e.g. by K1; a sample allowed to freely strain possessing an additional contribution  $\Delta K_1$  arising from the magnetoelastic energy. Even though a large number of materials possess relatively large magnetostrictions, only in a few materials, e.g. Tb<sub>3</sub>Fe<sub>2</sub>O<sub>1,2</sub>,  $\Delta$ K<sub>1</sub>>K<sub>1</sub>. As part of this paper we report the magnetic properties of Tb<sub>0,27</sub>Dy<sub>0,73</sub>Fe<sub>2</sub>. This pseudobinary compound, synthesized from highly magnetostrictive TbFe<sub>2</sub> and DyFe<sub>2</sub>, has the proper rare earth ratio to compensate the total magnetic anisotropy constant (K<sub>1</sub> tot) near room temperature while at the same time maintaining a large magnetostriction. Hence near room temperature,  $\Delta K_1$  and the intrinsic contribution  $K_1^{int}$  individually are large, but their sum  $K_1^{int}$  =  $K_1^{int}$  +  $\Delta K_1$  = 0.

## MAGNETIZATION AND MAGNETIC ANISOTROPY

The magnetizations of polycrystal TbFe  $_2$  , DyFe  $_2$  and TmFe  $_2$  have been known for some time.  $^{4,5,0}$  Measurements on single crystal Dyfe, at low temperatures were reported earlier. Because of the huge magnetic anisotropies of the RFe, compounds, magnetization measurements made on polycrystals using conventional laboratory fields do not yield the true saturation magnetic moments. Fields greater than 100 kOe are necessary to saturate the heavy rare earth compounds TbFe, through

TmFe<sub>2</sub>.
Single crystals of TbFe<sub>2</sub>, DyFe<sub>2</sub>, Tb<sub>0.27</sub>Dy<sub>0.73</sub>Fe<sub>2</sub> and TmFe<sub>2</sub> were grown by 0. D. McMasters by horizontal zoning and Czochralski techniques. The magnetic moments of these crystals were measured from 4 K to above their Curie temperatures (except for TbFe<sub>2</sub>) utilizing a vibrating sample magnetometer. In Fig. (1-4) the magnetic moments measured along their respective easy axes are compared with the earlier values of Burzo measured on polycrystals. In all cases, the moments of the single crystals are substantially higher. In the heavy RFe, series, the rare earth-iron exchange energy decreases with increasing rare earth atomic number approximately according to  $[(g-1)J(J+1)]^{1/2}$ . Thus for TmFe<sub>2</sub>, Thus for TmFe2, with a weak R-Fe exchange, the rare earth sublattice magnetization decreases rapidly with increasing temperature, falling below that of the iron sublattice near 235 K. For TbFe, and DyFe,, the exchange interaction is much stronger, leading to much higher moments, anisotropies and magnetostrictions at room temperature. In Table I we list the saturation values of the moment at 0 K and 300 K, the theoretical densities and the iron moments calculated assuming  $\mu_p = gJ\mu_h$ .

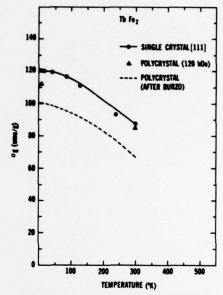


Fig. 1 Spontaneous magnetic moment of a TbFe single crystal as a function of temperature.

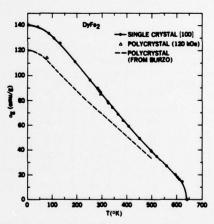


Fig. 2 Spontaneous magnetic moment of a DyFe2 single crystal as a function of temperature.

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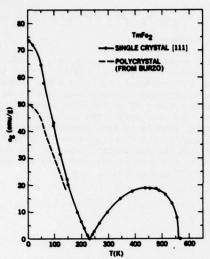


Fig. 3 Spontaneous magnetic moment of a TmFe<sub>2</sub> single crystal as a function of temperature.

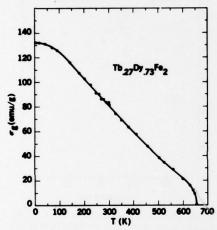


Fig. 4 Spontaneous magnetic moment of a Tb<sub>0 27</sub>Dy<sub>0 73</sub>Fe<sub>2</sub> single crystal as a function of temperature.

TABLE I. MAGNETIC MOMENTS, THEORETICAL DENSITIES AND CURIE TEMPERATURES OF RFe,

RFe <sub>2</sub>	σ(emu/g 0 K*	gm) 300 K	ρt gm/cm <sup>3</sup>	n(d) Fe 0 K	n <sub>b</sub> (e) Fe	T <sub>C</sub> K
TbFe,	120	88	9.06	1.60	1.75	697,711
DyFe,	140	87	9.28	1.57	1.60	635
HoFe,b	135	64	9.44	1.65	1.55	597,612
ErFe,c	116	29	9.62	1.61	1.60	590,597
TmFe <sub>2</sub>	74	10	9.79	1.64	1.55	560

Extrapolated

Taken from polycrystal data of K. H. J. Buschow and R. P. Van Stapele, J. de Physique 32, C1-672 (1971), and E. Burzo, Z. Angew, Phyzik 32, 127 (1971).

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Magnetic moment per iron atom in Bohr magnetons determined from Mössbauer spectra. W. E. Wallace, Prog. Rare Earth Sci. Tech. 3, 1 (1968).

For DyFe<sub>2</sub>, TmFe<sub>2</sub> and Tb<sub>0.27</sub>Dy<sub>0.73</sub>Fe<sub>2</sub>, the magnetic anisotropy constant K<sub>1</sub> was calculated from the fields required for magnetic saturation along the [100], [110] required for magnetic saturation along the [100], [110] and [111] directions. The anisotropy constants K<sub>1</sub> for DyFe<sub>2</sub> and TmFe<sub>2</sub> are plotted in Fig. (5). Except at the very low temperatures, the anisotropy of DyFe<sub>2</sub> is much larger than that of TmFe<sub>2</sub>. The single-ion theory has been successful in fitting the temperature dependence of the magnetostriction of the RFe<sub>2</sub> compounds as well as the anisotropy of ErFe<sub>2</sub>. Here we apply that theory of the magnetostriction of the kre<sub>2</sub> compounds as well as the anisotropy of ErFe<sub>2</sub>. Here we apply that theory to DyFe<sub>2</sub> and TmFe<sub>2</sub>. It is important to realize that even in the non-compensating RFe<sub>2</sub> compounds, because of the high magnetostrictions,  $\Delta K_1$  is not negligible. Taking the magnetoelastic contribution to be  $\Delta K_1 = -9c_4$ Taking the magnetoelastic contribution to be  $\Delta K_1 = -9c_1 + 2c_1/2$  and utilizing the single crystal magnetostriction data published earlier, we arrive at the values shown in Table II. The elastic constant  $c_1 = c_1/2$  to taken to be 4.87 x 10<sup>11</sup> dynes/cm for all compounds. From the measured values of  $K_1$  to the intrinsic anisotropy constants and  $\Delta K_1/K_1$  are calculated. According to single-ion theories, where J is large and the magnetically induced levels are assumed to be rearly evenly specific induced levels are assumed to be nearly evenly spaced, the expression for the temperature dependence of the anisotropy is given by:

$$K_1^{\text{int}}(T) = K_1^{\text{int}}(0) \hat{I}_{9/2}[\chi^{-1}(m_R(T))]$$

$$\Delta K_1(T) = \Delta K_1(0) \hat{I}_{5/2}^2 [ \chi^{-1}(m_R(T)) ].$$

Here the small temperature dependence of  $c_{44}$  is neglected.  $I_{2+1/2}$  is the hyperbolic Bessel function of order 2+1/2 normalized to one at T=0 K;  $\chi^{-1}$  is the inder k+1/2 normalized to one at k=0 k; k=1 is the inverse Langevin function and  $m_{\rm R}$  is the reduced rare earth sublattice magnetization. In Fig. (5) we show by the solid line the calculated anisotropy based upon  $k^{\rm int}$  (0) of 4.7 and -3.8 (x 10° erg/cm³) for DyFe<sub>2</sub> and TmFe<sub>2</sub> respectively. The values of  $\Delta K_1$  of -2.4° and -2.7° (x 10° erg/cm³) for DyFe<sub>2</sub> and TmFe<sub>2</sub> were taken from magnetostriction data. We find that with one adjustable constant per compound,  $k^{\rm int}_1$  (0), the fit to the data is within 20% over the temperature range of the experiments. within 20% over the temperature range of the experiments. For DyFe,, the Dy sublattice magnetization was taken from Mössbauer spectra by Bowden et al. For TmFe, the sublattice magnetization was calculated from the total magnetization reported in this paper less that of the iron sublattice inferred from Bowden et al. 12

TABLE II. MAGNETOELASTIC AND INTRINSIC CONTRIBUTION TO THE MAGNETIC ANISOTROPY (ergs/cm3).

RFe <sub>2</sub>	ΔK <sub>1</sub> (300)*	Kint (300)	$\Delta K_1(300)$
	x10 <sup>-4</sup>	x10 <sup>-4</sup>	$K_1^{int}(300)$
SmFe <sub>2</sub>	-970		
TbFe <sub>2</sub>	-1330	-6300	. 21
DyFe <sub>2</sub>	-350	2450	14
ErFe <sub>2</sub>	-20	-310	.06
TmFe <sub>2</sub>	-9.7	-43	.22

Calculated from  $\lambda_{11}$  values taken from A. E. Clark, J. R. Cullen, O. D. McMasters and E. R. Callen, AIP Conf. Proc. 29, 192 (1976) and R. Abbundi and A. E. Clark, J. Appl. Phys. 49, 1969 (1978).

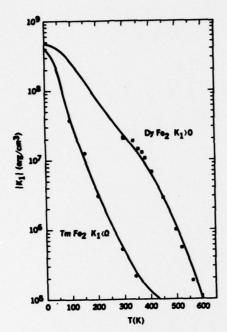


Fig. 5 Anisotropy constant K, for single crystals DyFe, and TmFe, as a function of temperature. The Solid curve represents the anisotropy calculated from single-ion theory.

The anisotropy constant  $K_1$  of  $Tb_{0.27}Dy_{0.73}Fe_2$  equals zero near room temperature.  $K_2$  remains negative. The anisotropy is  $DyFe_2$ -like ([100] easy) at low temperatures and  $TbFe_2$ -like ([111] easy) at high temperatures. The fields required for magnetic saturation along the principal directions are shown in Fig. (6). H<sub>111</sub>=H<sub>100</sub> at 283 K where H<sub>110</sub>=1.5 kOe. The anisotropy fields drop rather sharply from very high values at low temperatures. On the high temperature side, the slopes are not so steep, with the anisotropy constant K, reaching a peak value of -1.5 x 10 erg/cm at 380 K. The rapid drop of the anisotropy constants of both binary TbFe, and DyFe, over this temperature range accounts for this rather low peak value.

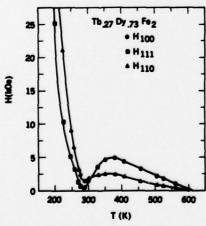


Fig. 6 Fields required for magnetic saturation along the principal crystallographic directions in single crystal Tb<sub>0.27</sub>Dy<sub>0.73</sub>Fe<sub>2</sub> as a function of temperature.

Magnetization rotation and domain wall motion can be effected at low applied magnetic fields when the magnetic anisotropy is low. The maximum energy transformed netic anisotropy is low. The maximum energy transformed from the magnetic to the elastic systems is  $\frac{1}{7}c_{44}\lambda_{111}$ . Defining  $f=\frac{1}{2}c_{44}\lambda_{111}/MH_{110}$  as a figure of merit of magnetostrictive transduction, we calculate for TbFe, f=0.016; and for DyFe, f=0.015. For the pseudobinary Tb<sub>0.27</sub>Dy<sub>0</sub>  $_{73}$ Fe, compound, f=0.5 with a peak near 283 K. In Fig. (7), we compare  $\frac{1}{2}c_{44}\lambda_{111}/MH_{110}$  to the relative magnetomechanical coupling factor k for temperatures near anisotropy compensation (maximum k  $\simeq 0.6$ ). Note that the slightly different pseudobinary composition (Tb<sub>0.26</sub>Dy<sub>0.74</sub>Fe<sub>2</sub>) used for the magnetomechanical coupling measurements shifts the compensating temperature to a little higher temperature. On the low temperature side of the anisotropy compensation, there is a rapid roll-off in both k and  $\frac{1}{2}c_{44}\lambda_{111}^{1}/M_{110}$ . At high temperature, the roll-off is not as severe, and in fact for k, rather small. The major source of the magnetostrains and the high constitutions. netostrains and the high coupling probably arises from domain wall motion rather than magnetization rotation. Above the anisotropy compensation temperature, the anisotropy remains low enough to prevent domain wall pinning at defect sites.

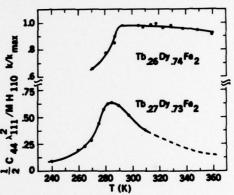


Fig. 7 Comparison of the relative magnetomechanical coupling factor for Tb  $_{2}$   $_{2}$   $_{2}$   $_{2}$   $_{2}$   $_{2}$   $_{3}$   $_{4}$   $_{111}$  /MH $_{110}$  (figure of merit for magnetostrictive transduction) for single crystal Tb<sub>0.27</sub>Dy<sub>0.73</sub>Fe<sub>2</sub> as a function of temperature.

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